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NEW DATA ON THE GEOCHEMISTRY OF INERT OR NOBLE GASES

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Having established (1) the accumulation of xenon heavy isotopes in ancient uranium minerals, and experimentally determined the xenon content of uranite from the Chernaya Salma deposit in North Karelia, we were able to propose and verify experimentally a new method and formula for determining the age of uranium minerals by the ratio of xenon to uranium in them. This formula has the following form:

$$t = \frac{1}{\lambda_\alpha} 2.30 \log_{10} \left[\frac{Xe}{0.14 N_1 (\lambda_\Д / \lambda_\alpha)} + 1 \right]$$

where

λ_α is the constant of the α-disintegration of uranium, equal to 1.52×10^{-10}/year;

Xe is the number of atoms of xenon found in a given amount of the mineral;

N_1 is the number of atoms of uranium found in a given amount of the mineral;

t is the geological age of the mineral in years;

$\lambda_\Д$ is the spontaneous fission constant of uranium, equal to 5.1×10^{-17}/year, according to N. A. Perfilov (2).

NOTE: Д is the initial letter of the Russian word for fission.

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Conversely, the same formula can be used to estimate how much xenon can accumulate in the earth's crust during the geological time of its existence. In this case, it is necessary to put the age of the earth's crust, 2×10^9 years, for t and the atomic Clarke (Numbers determining in percent the proportional content of a given element in a given cosmic body or part thereof) of uranium for N_1. Then the calculation will give the atomic Clarke of the xenon which can be formed by the process of spontaneous uranium fission in the earth's crust. The calculation shows that the quantity of xenon formed by this process in the earth's crust during the geological period of its existence is considerable and amounts to about 0.5 percent of the total xenon content of the earth's crust, including the atmosphere and the stratosphere, i.e., some tens of millions of cubic meters of xenon were thus formed. Such amounts of xenon must be considered in certain special cases of geochemistry.

Besides xenon, another inert gas, crypton, occurs in the earth's crust, due to spontaneous fission of uranium. However, the yield of crypton during uranium fission is considerably lower, 5 to 6 times, than that of xenon. Consequently, the total quantity of crypton which can accumulate in the earth's crust due to spontaneous disintegration of the uranium contained in it during the geological period of its existence must be smaller by a like amount, i.e., barely 1×10^7 m^3, which correspondingly constitutes hundredths of a percent of the total quantity of crypton contained in the earth's crust, including the atmosphere and stratosphere.

It must be added that it is very probable, although not yet proved experimentally in natural conditions, that both xenon and argon are also formed during the spontaneous fission of thorium and so in studying their geochemistry one must reckon with this supplementary source of their constant formation. However, since the spontaneous disintegration rate of thorium is almost one order below that of uranium, the amount of xenon and crypton being formed by this method also must be several times less than that formed by spontaneous fission of uranium and constitute only a comparatively small addition to the amounts formed by spontaneous fission of uranium. It is not yet possible to estimate these amounts with any degree of accuracy since the exact spontaneous fission constant of thorium is still unknown.

Thus, in the geochemistry of the noble gases, one must now reckon with natural nuclear processes other than the ordinary radioactivity which, as is well known, plays such an exceptional role in the geochemistry of helium. One of these processes is the process of the spontaneous fission of uranium and thorium, which results in the constant generation of two noble gases, xenon and crypton, in considerable quantities. In this connection, it must be emphasized that the isotopic composition of the noble gases formed in the earth's crust as a result of nuclear reactions differs from that of the noble gases of the atmosphere in all cases. For example, atmospheric helium and helium in natural gases contain, as was first shown by Alvarez and Cornog (3) in 1939, a very small amount of helium with a mass of 3, in addition to the helium isotope of mass 4. The content of the former was determined for atmospheric helium by Alvarez and Cornog as $He^3/He^4 = 1 \times 10^{-7}$, and for helium in natural gas as $He^3/He^4 = 1 \times 10^{-8}$.

In 1946 at the Radium Institute, employing as did Alvarez and Cornog a cyclotron as a mass-spectrograph and using thick-layer photographic plates for registering particles with masses of 3 and 4, M. Meshcheryakov, Ye. Grigor'yev, A. Reut and T. Ehrenin established that in helium isolated from uranite by E. K. Gerling, there was no helium with a mass of 3 or, more accurately, that its content in helium of radioactive origin is in any case at least 5,000 times less than in helium from the atmosphere, i.e., $He^3/He^4 < 10^{-10}$ for helium of radioactive origin.

At the same time the value $He^3/He^4 = 6 \times 10^{-8}$ was determined for the He^3 content in helium from the natural gas escape of the Mel'nikovo deposit, taking the value of He^3/He^4 for the atmosphere as 1×10^{-7} in agreement with Alvarez and Cornog. However, taking Nier's (4) newer and more accurate figure,

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He^3/He^4 = 1 x 10^{-6} (He^3/He^4 = 9 x 10^{-5}), for the atmosphere, there is a corresponding alteration, i.e., the content of He^3 in natural gas is increased.

Of great interest is the discovery made at the Radium Institute in 1946, that helium of radioactive origin and helium from subterranean gaseous streams have completely different isotopic compositions. It compels us to examine anew the concepts of the genesis of subterranean helium which have been established at the present time. All of us at the present time accept the fact that natural-gas helium is of radioactive origin. Now, on the basis of the works of M. G. Meshcheryakov and his colleagues and of E. K. Gerling and M. G. Meshcheryakov, carried out at the Radium Institute in 1946, we have for the first time experimental proof of the hypothesis previously accepted by Moureu (5) concerning the dual origin of helium, that natural-gas helium is a mixture of helium of radioactive origin and helium of some other origin, relatively rich in the isotope with a mass of 3. This follows from the fact that the content of the mass 3 isotope in natural-gas helium is slightly lower than its content in air and, conversely, for the ratio:

$$\frac{He^2/N_2 \text{ in natural gases}}{He^3/N_2 \text{ in the atmosphere}}$$

we obtain a value far greater than one. In point of fact, this ratio for the Mel'nikovskiy deposit is 230, and for the gas of American deposits from 1200 to 1,400 while for the ratio:

$$\frac{Ar/N_2 \text{ in natural gases}}{Ar/N_2 \text{ in the atmosphere}}$$

we always have a value close to one. This shows that the argon and nitrogen in natural-gas streams are usually of atmospheric origin, while helium with a mass of 3 is of nonatmospheric origin. Thus, the new data on the isotopic composition of natural-gas helium again compels us to recall Moureu's hypothesis, stating in effect that noble gases and, in particular, subterranean helium are of primary origin or, as he expresses it, astrophysical origin, i.e., they were occluded by our planet during its formation from the primary gaseous atmosphere. The new data furnishes for the first time some experimental bases for such an hypothesis, with the essential correction that not all but a certain part of the subterranean helium is of primary origin in accordance with Moureu's hypothesis. Bessel's calculations would also support this idea, since he showed that the average period for which an atom of helium with a mass of 4 remains in the atmosphere before escaping is 3 x 10^7 years and so the main part of the He^3 of the atmosphere must have escaped during the geological period of the earth's existence. One must suppose that the atmosphere a few hundred million years ago was far richer in the helium isotope with mass 3 than is now the case; and 2 billion years ago, even more so. If, therefore, during remote geological periods of the earth's existence, helium from the atmosphere was mixed with helium of radioactive origin, due to processes in the earth's crust, such a mixture might have an isotopic composition corresponding to that of helium from natural-gas streams at the present time.

However, in addition to the supposition just stated concerning the genesis of a part of the helium contained in subterranean helium, other theories can be advanced. In particular, it is possible to suggest that the helium with mass 3 is itself a product of some nuclear reactions which took place earlier, or are even still in progress in the earth's crust. At the present time, such nuclear reactions are unknown to us; however, only further experimental investigations can solve the problem which of the two suppositions, concerning the genesis of the helium isotope with mass 3 in subterranean helium, is to be preferred.

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The great interest of the recent experiments performed at the Radium Institute to which we have made reference lies in the fact that they compel us to re-examine the problem of the origin of subterranean gases and for the first time, or so it seems to us, convincingly demonstrate the importance of studying the geochemistry, not merely of the individual elements, but also of their individual isotopes, since only such a study can allow the part they play in the geochemistry of the elements to be accurately evaluated and correctly understood.

In conclusion, we would like to dwell a little on the fact that of recent years work is being done on the nuclear reactions of argon, a new advance in the geochemistry of yet another noble gas. It appears that this element also can be formed during certain nuclear reactions. This conjecture, expressed as far back as 1937, was proved experimentally by Thomson and Rowlands (6), who showed that potassium with mass 40, in addition to its β -radioactivity, which has been known for a long time, undergoes other radioactive transformations by the capture of an electron from the K-electronic shell by the nucleus of the atom. When this occurs, an argon isotope with mass 40 is formed from potassium of atomic weight 40. The authors also calculated the probability of such a process, compared with the well-known conversion process $K \rightarrow Ca$. These calculations, taking into account the high value of K-radiation in the salt itself, cannot possibly be of outstanding accuracy. Nevertheless, the calculation showed that for one β-transformation there are approximately three K-capture transformations. On the basis of this calculation, we obtain the value 1.8×10^9 years for T, the half-life of K into Ar with a mass of 40 . We recall that the half-life T for the β-disintegration of potassium 40 is 1.8×10^9 years. If the K-capture probability of potassium 40 were really as great as follows from the calculations of Thomson and Rowlands, then enormous quantities of argon would have been formed in this manner during the geological time of existence of the earth's crust, and the role of this process in the geochemistry would have been very great.

As yet no experiments are known which would show and, still more, establish quantitatively the accumulation of argon with mass 40 in natural objects. We can, however, even now assert that although the process of radioactive transformation of calcium 40 by K-capture does take place, as was convincingly demonstrated by Thomson and Rowlands, the probability of the process is far from being as great as calculated by these authors. This may be said to follow from A. A. Cherepenikov's (7) investigation of the gases contained in the potassium minerals of the Solikamsk deposit. These gases are not perceptibly enriched by argon, which would have been expected if the calculations of Thomson and Rowlands had been correct.

Of prime importance now is the accomplishment of systematic quantitative investigations on the composition of the gases evolved from minerals rich in potassium and of various geologic ages. The object is to furnish conclusive proof of the accumulation of argon, and especially argon with a mass of 40, in them during the passage of time and thus enable the probability of the K-capture process in potassium to be determined by another method and the importance of the role of this process in the geochemistry of argon to be evaluated. At the same time, investigations of this kind would lead to an independent method of determining the absolute geologic age of natural formations.

Such investigations are now in progress in the gas laboratory of the Radium Institute of the Academy of Sciences USSR.

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